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著者(和文)	LuoWeikai
Author(English)	Weikai Luo
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Thesis Outline

The structural and magnetic characterizations of FeCoNiAlCr HEA films have been studied in this thesis. In this work, the FeCoNiAlCr HEA films with different compositions are prepared on the quartz glass substrate by DC reactive magnetron sputtering. The microstructure and magnetic, electrical, and mechanical properties were investigated. Annealing is used as the main treatment to achieve the adjustment of the magnetic properties of the films, due to the microstructure change and element migration during the annealing process. Then, Al content, which can introduce crystal structure change to the films, was chosen as the variable. The effect of the Al content on magnetic properties and the mechanism were studied. Finally, by introducing nitrogen to the system, FeCoNiAlCr N-HEA films were prepared. The effect of nitrogen incorporation on microstructure and magnetic properties was studied. The main conclusions and detailed experiment results are summarized as follows:

i. **Deposition of HEA films on the glass substrate**

The FeCoNiAlCr thin films can be prepared by magnetron sputtering method using independent bulk pure metal as the target. The composition of films can be controlled by changing the relative area of each target. Due to the sputtering rate differing from targets and position, the area of the target is not proportional to the composition percentage of the film. Thus, certain lay-out method of targets corresponds to certain compositions. And the compositions need to be confirmed by ICP-OES.

ii. **$Fe_{0.18}Co_{0.36}Ni_{0.21}Al_{0.10}Cr_{0.15}$ of HEA films with soft magnetic properties**

The as-deposited $Fe_{0.18}Co_{0.36}Ni_{0.21}Al_{0.10}Cr_{0.15}$ HEA films have an FCC structure, which is

consistent with the VEC criterion (VEC=7.98). With the increase of annealing temperature, the intensity of the peak increases, but there is no obvious change in the peak position, indicating that the crystallinity becomes better, but there is no phase transformation.

The soft magnetic properties are evaluated of $Fe_{0.18}Co_{0.36}Ni_{0.21}Al_{0.10}Cr_{0.15}$ thin films exhibit a dynamic magnetic evolution as the annealing temperature changes, In this study, the soft magnetic properties are mainly evaluated by Ms, Hc and resistivity. The annealing treatment can effectively improve the Ms within a certain temperature range (400-800 °C) due to the segregation of antiferromagnetic element Cr after annealing. The Hc shows nonlinear change due to the grain growth during annealing. The resistivity and mechanical properties are thermally stable, and the influence of the annealing process is almost negligible up to 800 °C.

The optimal properties can be obtained after annealing under 800 °C. At this time, $Fe_{0.18}Co_{0.36}Ni_{0.21}Al_{0.10}Cr_{0.15}$ HEA films have high Ms (698 emu/cm^3), moderate Hc (87 Oe), high resistivity (110~120 $\mu\Omega \cdot cm$), good mechanical properties, and good thermal stability, which can be applied as functional soft magnetic thin films with outstanding performance.

iii. Effect of Al content on microstructure and magnetic properties of FeCoNiAlxCr HEA films

Al, as a major alloying element in most of the HEAs studied to date, could lead to the crystal structure transformation from FCC to BCC. In this study, the crystal structure of FeCoNiAlxCr films can transform from FCC (x=0.10, 0.14) to BCC (x=0.10, 0.14). For the FCC FeCoNiAlxCr films (x=0.10, 0.14), with the increase of annealing temperature, the crystallinity becomes better, but there is no phase transformation. For the BCC FeCoNiAlxCr films, with the increase of annealing temperature, the peak intensity increase may also be due to the

appearance of some new phase.

The magnetic measurements show that the as-deposited FCC (x=0.10, 0.14) films can show soft magnetism, and BCC (x=0.10, 0.14) films show poor magnetism. After the annealing process, all the FeCoNiAlxCr films can show soft magnetism and in-plane anisotropy. The $FeCoNiAl_{0.14}Cr$ HEA films annealed at 700 °C show the best soft magnetism. At this time, Ms is 996 emu/cm^3, and Hc is 105 Oe. For as-deposited films, the effect of Al content on Ms is limited. The increase of the Al component can refine the grain and increase the Hc.

During the annealing process, the segregation behavior of elements can explain the change of microstructure and the change of Ms. The Al element plays an important role in the segregation behavior of elements. When the ratio of Al increased from 10 at.% to 32 at.%, the Fe, Co, and Ni elements were easier to segregate. Al element matrix acts as a sort of migration channel that facilitates the segregation of the Fe/Co/Cr, especially Cr elements, which in turn affects the Ms of FeCoNiAlxCr HEA films.

Increasing the annealing temperature facilitates the growth of grains, which is the main reason causing the change of coercivity. Also, the annealing can release stress and decrease grain boundaries. These changes can comprehensively influence the Hc.

The FeCoNiAlxCr films also show high resistivity. When the Al component increase, the resistivity increase.

Overall, the optimal magnetic properties can be obtained for FeCoNiAl0.14Cr films after annealing at 700 °C.

iv. **Effect of nitrogen incorporating on microstructure and magnetic properties of FeCoNiAlCr HEA films**

The magnetic properties and microstructure of $FeCoNiAl_{0.14}Cr$ N-HEA films have been investigated. The proportion of nitrogen is adjustable by controlling the flow rate of nitrogen during the magnetron sputtering process.

The as-deposited film has an amorphous-like structure, which is due to the nitrogen lead high mixing entropy and large atomic size difference. The as-deposited films show ultra-good soft magnetism. Because the combination between N and Cr can reduce the influence on the magnetic elements.

The films were annealed at 400-800 °C. The annealing treatment can effectively improve the Ms within a certain temperature range (400-700 °C) due to the segregation of magnetic elements after annealing. The phase transition from amorphous to partial FCC also is observed after annealing.

In summary, during the annealing process, because nitrogen stabilizes the microstructure and tends to bind with Al and Cr, Al and Cr are localized by N, and relatively Fe/Co/Ni may segregate first. The higher the proportion of nitrogen is, the easier the segregation happens. The segregated Fe-Co-Ni phase enhances the Ms.

v. **Optimal properties**

Overall, the optimal properties can be obtained for the as-deposited FeCoNiAl0.14Cr N-HEA films. At this time, nitrogen incorporating FeCoNiAl0.14Cr HEA films have high Ms (1147 emu/cm3), not too high Hc (75Oe), high resistivity (153 μΩ·cm), and commendable mechanical properties. This combination of characteristics makes them suitable for application as high-performance functional soft magnetic thin films.